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Light Scattering from an Atom Near the Surface of a Superlattice

by

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Light scattering from an atom near the surface of a superlattice

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Abstract

A semiclassical theory is developed to treat the interaction of radiation with an atom located near the surface of an m-component superlattice. For the special case of $m = 2$, the radiation penetration depth into the superlattice and the resonance fluorescence spectrum are calculated numerically for superlattices of metal-insulator construction. The results show sensitive dependence on the plasma frequency in the metal layer as well as on the dielectric function and width of the insulator layer.

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I. Introduction

When an atom or molecule near or adsorbed on a solid surface interacts with a radiation field, the presence of the surface and substrate greatly influences the optical phenomena. Examples include surface-enhanced Raman scattering,[1,2] coherence and energy transfer in spontaneous emission,[3-6] surface-induced resonance fluorescence,[7-13] and so on. The optical properties of adspecies on a solid surface may provide a sensitive probe of electronic and other structure of the substrate, and hence has prompted a careful re-examination of the optics of surfaces. Once the mechanism of these optical processes is fully understood and brought under experimental control, it should become a powerful tool for the analysis of surface processes.

On the other hand, when a strong driving coherent field is nearly on resonance with the adatomic transition frequency, the field places the driven atom in an environment in which the probability of stimulated emission may exceed that of spontaneous emission. When this is the case, ac Stark splitting of the atomic levels, Rabi oscillations of the level occupation probabilities and nutational oscillations of the stimulated field intensity are enhanced. Consequently, resonance fluorescence[7-14] and other nonlinear optical phenomena become possible.[15,16] Because multiphoton transitions are just as important as the single-photon transition in such light-driving processes, the ordinary perturbation method is no longer reliable.

In the surface-free case such as atoms in a gas,[15] such processes can be effectively dealth with by the well-known optical Bloch equation (OBE). With the presence of the surface, a set of surface-dressed optical Bloch equations (SBE) has been derived[7-10] to treat such problems as the effects of surface-reflected photons, the resonance interaction of an adatom with surface plasmons, collisional dephasing of the adatom due to the gas atoms in the medium, and random phase fluctuation of the intense laser field. Resonance fluorescence has been investigated for an adatom near a flat metallic surface[7,8] and near a rough metallic surface represented by a hemispheroid protrusion on a flat surface.[9,10] More recently, a different approach involving reservoir theory[17] and Dekkars's quantization procedure for a dissipative system[18,19] has been adopted to derive another set of the SBE.[11] The resonance fluorescence spectrum of an adatom has been analyzed for the following cases: an adatom is considered near the surface of a metal sphere with the size of the sphere taken into account,[12] and the problem is considered again near the surface of a semi-infinite conductor and dielectric superlattice in which the frequency of the transverse optical phonon matches that of the adatomic transition.[11]

We investigate, in this paper, the resonance fluorescence spectrum of an adatom near the surface of a superlattice. The adatom is taken as an emitting dipole. The emitted field is reflected back from the surface and interfaces and is coupled to the radiating dipole whose dynamical behavior is therefore totally changed. We first develop the theory for a general superlattice composed of m different layers per period. This theory may be applied to simulate quasi-periodic systems.[20,21] Numerical results are given for the special case of m = 2.

II. Theory

For a two-level atom driven by a monochromatic laser field with

$$E(t) = \frac{1}{2} (Ee^{i\omega_0 t} + E^* e^{-i\omega_0 t}) \quad , \tag{1}$$

the SBE are given by[12-14]

$$\frac{d}{dt}\begin{bmatrix} <S^+> \\ <S^z> \\ <S^-> \end{bmatrix} = \begin{bmatrix} i(\Delta+\Omega^s) - \gamma & i\Omega & 0 \\ i\Omega^*/2 & -2\gamma & -i\Omega/2 \\ 0 & -i\Omega^* & -i(\Delta+\Omega^s) - \gamma \end{bmatrix} \begin{bmatrix} <S^+> \\ <S^z> \\ <S^-> \end{bmatrix} - \begin{bmatrix} 0 \\ \gamma \\ 0 \end{bmatrix} . \quad (2)$$

The notation is as follows. The adatom with a transition frequency ω is located at a distance d_o away from the surface of the superlattice. The matrix element of the electric dipole moment operator is denoted by $|p|$, and E and ω_o are the amplitude and frequency of the external laser field, respectively. The detuning is $\Delta = \omega - \omega_o$, and the Rabi freuquency is $\Omega = |p|E$. The transition probability amplitude is proportional to the projection operators defined by

$$S^+ = |+><-|$$

$$S^z = \frac{1}{2}(|+><+| - |-><-|) \quad (3)$$

$$S^- = |-><+| \quad .$$

The total decay rate of the adatom can be written as

$$\gamma = \gamma^o + \gamma^s \quad , \quad (4)$$

where γ^o is the decay rate in the absence of the substrate,[22]

$$\gamma^o = \frac{2}{3}\sqrt{\epsilon_o}\,|p|^2\omega^3/c^3 \quad , \quad (4a)$$

and

$$\gamma^s = |p|^2 \ \mathrm{Im}\ f(d_o) \tag{4b}$$

is the decay rate induced by the surface. The frequency shift of the spontaneous radiation due to the surface is

$$\Omega^s = |p|^2 \ \mathrm{Re}\ f(d_o) \quad , \tag{5}$$

and the function $f(d_o)$ is determined by[11,13,14]

$$E_R = |p| f(d_o) S^- = p f(d_o) \quad , \tag{6}$$

where E_R is the component of the reflected field $\vec{E}_R$ in the direction of $\vec{p}$.

Equation (2) agrees with the result from linear response theory[6] when the adatom is taken as a harmonic oscillator. With the aid of the regression theorem for correlation functions,[23,24] we find from the SBE the well-known results of the incoherent resonance fluorescence spectrum,[12-14]

$$\phi_{inc}(\nu) = \frac{1}{2} |\Omega|^4 \gamma (D^2 + \frac{1}{2} |\Omega|^2 + 4\gamma^2)/(\frac{1}{2}|\Omega|^2 + |z|^2)(x^2+y^2) \quad , \tag{7}$$

where $D = \nu - \omega_o$, $z = \gamma + i(\Delta+\Omega^s)$, $x = 2\gamma(\frac{1}{2}|\Omega|^2 + |z|^2 - 2D^2)$, and $y = D(|\Omega|^2 + |z|^2 + 4\gamma^2 - D^2)$.

Consider now a semi-infinite superlattice consisting of m different layers per period as shown in Fig. 1. The surface is taken to be the xy-plane, and an adatom with dipole moment $\vec{p}$ is located at $\vec{r} = \vec{r}_o = (0,0,-d_o)$. The dielectric constants and thicknesses of these layers are ϵ_i and d_i, respectively, where $i = 1, 2, \ldots, m$. The reflected electric field at r_o from a substrate of a two-component superlattice has been worked out in Ref. 8. We now generalize the theory to the case of an m-component superlattice in which the thickness of each period is L. The set of equations for the fields $\vec{E}$ and $\vec{H}$ in different materials can be obtained directly from Maxwell's equations. The results are[6,11,13,14]

$$\nabla^2\vec{E} + \epsilon_o k_o^2\vec{E} = -4\pi[k_o^2\vec{P} + \frac{1}{\epsilon_o}\nabla(\nabla\cdot\vec{P})]$$

$$\vec{H} = \frac{1}{ik_o}\nabla\times\vec{E} \quad , \qquad z < 0 \tag{8a}$$

$$\nabla^2\vec{E}_1 + \epsilon_1 k_o^2\vec{E}_1 = 0 \quad , \qquad \nabla\cdot\vec{E}_1 = 0$$

$$\vec{H}_1 = \frac{1}{ik_o}\nabla\times\vec{E}_1 \quad , \qquad nL \le z \le nL + d_1 \tag{8b}$$

$$\nabla^2\vec{E}_2 + \epsilon_2 k_o^2\vec{E}_2 = 0 \quad , \qquad \nabla\cdot\vec{E}_2 = 0$$

$$\vec{H}_2 = \frac{1}{ik_1}\nabla\times\vec{E}_2 \quad , \qquad nL + d_1 \le z \le nL + d_1 + d_2 \tag{8c}$$

$$\nabla^2\vec{E}_m + \epsilon_m k_o^2\vec{E}_m = 0 \quad , \qquad \nabla\cdot\vec{E}_m = 0$$

$$\vec{H}_m = \frac{1}{ik_o}\nabla\times\vec{E}_m \quad , \qquad (n+1)L - d_m \le z \le (n+1)L \quad , \tag{8d}$$

where $k_o = \omega/c$ is the wave number in vacuum, n labels the period in the superlattice, and $\vec{P}$ is defined by

$$\vec{P}(\vec{r},\omega) = \vec{p}(\omega)\ \delta(\vec{r}-\vec{r}_o) \quad . \tag{9}$$

The solution for the electric field in Eq. (8a) can be written as

$$\vec{E}(\vec{r}\,,w) = \int du \int dv\ \vec{\mathcal{E}}(u,v,\omega)\ e^{i\vec{k}'\cdot\vec{r}} + \vec{E}_p(\vec{r},\omega) \quad , \qquad z < 0$$

$$\vec{k}' \cdot \vec{\mathcal{E}} = 0 \quad , \qquad \vec{k}' = (k_{\parallel}, -w) \quad , \qquad w^2 = \epsilon_o k_o^2 - k_{\parallel}^2 \quad , \qquad \operatorname{Im} W \geq 0 \tag{10}$$

where we have defined $\vec{k} = (\vec{k}_{\parallel}, w) = (u,\ v,\ w)$. The field due to the dipole is

$$\vec{E}_p(\vec{r},\omega) = -\frac{1}{2\pi i}\int du \int dv\ \frac{k_o^2\vec{p} - \epsilon_o^{-1}\nabla(\vec{p}\cdot\nabla)}{w}$$

$$\times \exp[iu(x-x_o) + iv(y-y_o) + iw|z-z_o|] \quad . \tag{11}$$

Because of the periodic structure of semi-infinite superlattice, the electric field may have a Bloch-wave-like form with an envelope function decaying exponentially with increasing z. Thus we find solutions for the electric field of Eqs. (8b)-(8d)[13,25] as

$$\left.\begin{aligned} \vec{E}_1(\vec{r},\omega) &= \int du \int dv \left\{ \vec{\mathcal{E}}_1^{(+)}(u,v,\omega) e^{iw_1(z-nL)} \right. \\ &\quad \left. + \vec{\mathcal{E}}_1^{(-)}(u,v,\omega)\ e^{-iw_1(z-nL)} \right\} e^{i\vec{k}_{\parallel}\cdot\vec{r} - \beta nL} \\ \vec{k}_1 \cdot \vec{\mathcal{E}}_1^{(+)} &= \vec{k}_1' \cdot \vec{\mathcal{E}}_1^{(-)} = 0 \end{aligned}\right\} \tag{12a}$$

$$\left.\begin{aligned}
&\vec{k}_1 = (\vec{k}_\parallel, w_1) \quad , \qquad \vec{k}_1' = (\vec{k}_\parallel, -w_1) \\
&w_1^2 = \epsilon_1 k_o^2 - k_\parallel^2 \quad , \qquad nL \le z \le nL + d_1
\end{aligned}\right)$$

$$\left.\begin{aligned}
&\vec{E}_2(\vec{r},w) = \int du \int dv \left\{ \vec{\mathcal{E}}_2^{(+)}(u,v,w)\ e^{iw_2(z-nL-d_1)} \right. \\
&\qquad \left. + \vec{\mathcal{E}}_2^{(-)}(u,v,\omega) e^{-iw_2(z-nL-d_1)} \right\} e^{i\vec{k}_\parallel\cdot\vec{r} - \beta nL} \\
&\vec{k}_2 \cdot \vec{\mathcal{E}}_2^{(+)} = \vec{k}_2 \cdot \vec{\mathcal{E}}_2^{(-)} = 0 \\
&\vec{k}_2 = (\vec{k}_\parallel, w_2) \quad , \qquad \vec{k}_2' = (\vec{k}_\parallel, -w_2) \\
&w_2^2 = \epsilon_2 k_o^2 - k_\parallel^2 \quad , \qquad nL + d_1 \le z \le nL + d_1 + d_2
\end{aligned}\right\} \tag{12b}$$

$$\left.\begin{aligned}
&\vec{E}_m(\vec{r},\omega) = \int du \int dv \left\{ \vec{\mathcal{E}}_m^{(+)}(u,v,\omega)\ e^{iw_m[z - (n+1)L + d_m]} \right. \\
&\qquad \left. + \vec{\mathcal{E}}_m^{(-)}(u,v,\omega)\ e^{-iw_m[z - (n+1)L + d_m]} \right\} e^{i\vec{k}_\parallel\cdot\vec{r} - \beta nL} \\
&\vec{k}_m \cdot \vec{\mathcal{E}}_m^{(+)} = k_m \cdot \vec{\mathcal{E}}_m^{(-)} = 0 \\
&\vec{k}_m = (\vec{k}_\parallel, W_m) \quad , \qquad \vec{k}_m' = (\vec{k}_\parallel, -w_m) \\
&w_m^2 = \epsilon_m k_o^2 - k_\parallel^2 \quad , \qquad (n+1)L - d_m \le z \le (n+1)L
\end{aligned}\right\} \tag{12c}$$

where we have introduced a parameter β to measure the rate of absorption.

For simplicity, we assume that the dipole is orientated perpendicular to the interfaces of the superlattice. We can then follow the standard procedure to find the component of the reflected field along the dipole direction at $\vec{r}_o$. Equations (12) together with the usual Maxwell boundary conditions at the surface and interfaces then yield

$$E_R = \frac{i}{\epsilon_o} k_o^3 \, p \, \phi(d_o) \tag{13}$$

$$\phi(d_o) = \int_0^\infty \frac{k^3 dk}{U_o} e^{2iU_o \hat{d}_o} \psi_1/\psi_2 \tag{14a}$$

$$\begin{aligned} \psi_1 = &- 2^{m-1} (1 - V_{10}) e^{\beta L} \\ &+ \sum_{N_m=0}^{1} \Big\{ [1 - (-1)^{N_m} V_{mo}] \sum_{N_2,\ldots,N_{m-1}=0}^{1}{}' \Big\{ \exp[i \sum_{j=1}^{m} (-1)^{N_j} U_j \hat{d}_j] \\ &\times \prod_{\ell=1}^{m-1} [1 + (-1)^{N_\ell + N_{\ell+1}} V_{\ell,\ell+1}] \Big\}\Big\} \end{aligned} \tag{14b}$$

$$\begin{aligned} \psi_2 = &\; 2^{m-1}(1 + V_{10}) e^{\beta L} \\ &- \sum_{N_m=0}^{1} \Big\{ [1 + (-1)^{N_m} V_{mo}] \sum_{N_2,\ldots,N_{m-1}=0}^{1}{}' \Big\{ \exp[i \sum_{j=1}^{m} (-1)^{N_j} U_j \hat{d}_j] \\ &\times \prod_{\ell=1}^{m-1} [1 + (-1)^{N_\ell + N_{\ell+1}} V_{\ell,\ell+1}] \Big\}\Big\} \quad , \end{aligned} \tag{14c}$$

where $N_1 = N_{m+1} = 0$, $V_{n\ell} = \epsilon_n U_\ell / \epsilon_\ell U_n$, $U_\ell = \sqrt{\epsilon_\ell - k^2}$, $\hat{d}_\ell = k_o d_\ell$, and

$n = 1, 2, \ldots, m$. Because of the periodicity of the superlattice, we define $v_{m,m+1} \equiv v_{m,1}$. The symbol $\sum'$ means summation when $m \geq 3$ and no summation when $m = 2$. The attenuation factor $e^{\beta L}$ is determined by the equations

$$e^{\beta L} = \frac{1}{2} [\eta \pm \sqrt{\eta^2 - 4}] \quad , \qquad \mathrm{Re}\beta \geq 0 \tag{15}$$

$$\eta = 2^{1-m} \sum_{N_2 \ldots N_m = 0}^{1} \left\{ \cos[\sum_{j=1}^{m} (-1)^{N_j} U_j \hat{d}_j] \prod_{\ell=1}^{m} [1 + (-1)^{N_\ell + N_{\ell+1}} V_{\ell,\ell+1}] \right\} . \tag{16}$$

If the superlattice is composed of highly-dissipative materials, the incident electric field cannot go very deep into the layers. When the condition $L << \lambda$ is satisfied, which is usually the case, we can define the penetration depth for the field as

$$\hat{d}_p = \alpha L = (\mathrm{Re}\beta)^{-1} \quad , \tag{17}$$

where α is a positive number.

It is observed from the above equations that both the reflected field and the penetration depth depend strongly upon the number of layers per period as well as the thickness and dielectric properties of the individual layer. Meanwhile, the surface plasmon TM modes are determined by the zeroes of the denominator, namely, $\psi_2 = 0$. The properties of the surface plasmon as well as the interference between the electric fields reflected and refracted at each interface play very important roles in the determination of E_R, both its amplitude and its phase.

To find the spontaneous decay rate and frequency shift, we first note that a comparison of (6) and (13) yields $f(d_o) = \frac{i}{\epsilon_o} k_o^3 \phi(d_o)$. Substituting into (4) and (5), we obtain

$$\gamma = 1 + \frac{3}{2} \epsilon_o^{-3/2} \operatorname{Re} \phi(d_o) \tag{18a}$$

$$\Omega^s = - \frac{3}{2} \epsilon_o^{-3/2} \operatorname{Im} \phi(d_o) \tag{18b}$$

expressed in the unit of γ^o.

III. Numerical results

The theory developed above is valid for any number of layers in one period of the superlattice. In what follows, we shall apply it to a typical case of two-component superlattices. The first layer in each period is a metal with dielectric function $\epsilon_1(\omega) = 1 - \omega_p^2/(\omega^2 + i\omega\gamma_p)$, while the other is an insulator with a wide energy gap. Setting $m = 2$ in Eqs. (14b), (14c) and (16), we have

$$\psi_1 = -2(1-V_{10})\, e^{\beta L} + [(1-V_{20})(1+V_{12})e^{iU_2\hat{d}_2} + (1+V_{20})(1-V_{12})\, e^{-iU_2\hat{d}_2}]e^{iU_1\hat{d}_1} \tag{19a}$$

$$\psi_2 = 2(1+V_{10})e^{\beta L} - [(1+V_{20})(1+V_{12})e^{iU_2\hat{d}_2}$$

$$+ (1-V_{20})(1-V_{12})e^{-iU_2\hat{d}_2}]\, e^{iU_1\hat{d}_1} \tag{19b}$$

$$\eta = \frac{1}{2}\Big[(1+V_{12})(1+V_{21})\cos(U_1\hat{d}_1 + U_2\hat{d}_2)$$

$$+ (1-V_{12})(1-V_{21})\cos(U_1\hat{d}_1 - U_2\hat{d}_2)\Big] \quad . \tag{20}$$

We look at the penetration depth for normal incidence of light with frequency ω. From (15), (17) and (20), we calculate $\hat{d}_p$ as a function of the adatomic transition frequency. The results for different layer thicknesses are shown in Fig. 2, in which we have defined $x = \omega/\omega_p$. It is observed that for fixed ω, the penetration depth has a sensitive dependence on the charge carrier density in the metal layers. It decreases with increasing carrier density, which is easy to understand because a large carrier density results in more collisions and therefore more energy loss of the incident radiation. Furthermore, we also see that the larger the fraction that the nonabsorptive insulator in each period occupies, the deeper the field penetrates into the superlattice.

The incoherent part of the resonance fluorescence spectrum may be investigated numerically. We find again that the spectrum is extremely sensitive to the plasma frequency or the density of charge carriers in the metallic layers. When ω is fixed, different carrier densities may result in spectra with different number and shape (height and width) of peaks as well as the position of sidebands. The situation is illustrated in Fig. 3 in which the spectrum is shown as a function of D [defined just beneath Eq. (7)] for various values of x. That the sidebands do not appear in the case of the one-peak spectrum may be understood as follows.

When x is such that ψ_2 is near its minimum, the resonance condition for the surface plasmon to exist is nearly satisfied. The creation of the surface plasmon then enhances the absorption and results in a single-peak spectrum. This is because non-radiative energy transfer to the surface plasmon becomes very strong near resonance and hence greatly increases the absorption of the incident energy by the surface via the adatom. Consequently, the scattered field intensity is too weak to produce the sidebands of the spectrum. On the other hand, when x is such that ψ_2 is far from its minimum, this absorption process is insignificant, and the spectrum has three peaks which are all high and narrow, provided that there is no significant absorption resulting from the multibeam interference.

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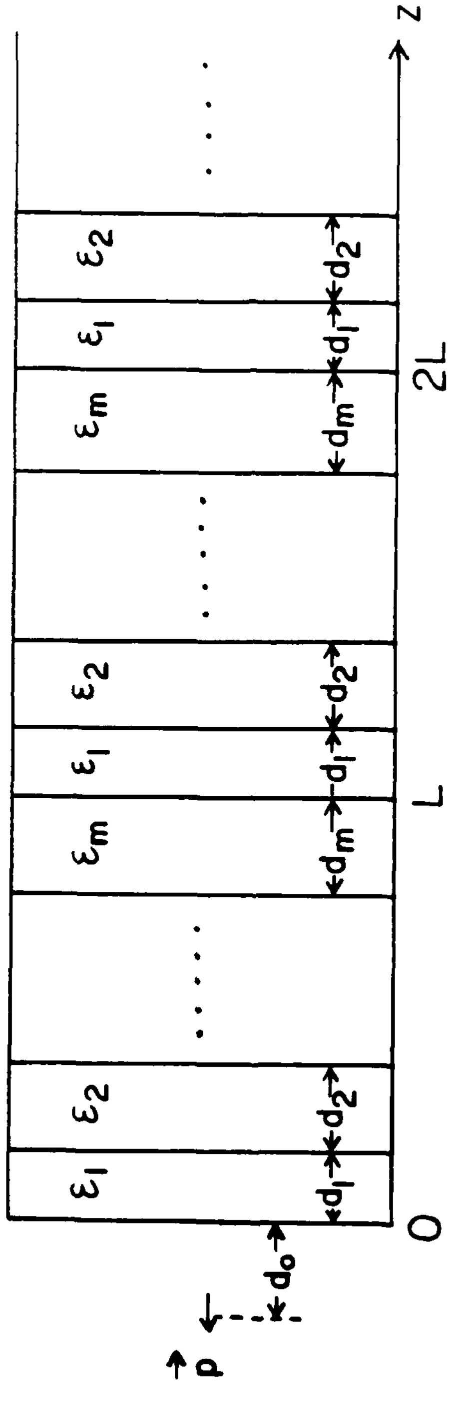
Figure Captions

1. Schematic diagram of a semi-infinite superlattice with period L. An atomic dipole moment is located in front of the surface.

2. Penetration depth as a function of x for a metal-insulator two-component superlattice. The parameters employed are: $\gamma_p = 0.01\ \omega_p$, $\epsilon_2 = 0.3$, $d_1 = 0.1$ and (a) $d_1 = 0.5$, (b) $d_2 = 0.2$.

3. Incoherent resonance fluorescence spectrum from a two-component metal-insulator superlattice for different x values. The parameters are: $\Delta = 2.0$, $|\Omega| = 45$, $\gamma_p = 0.01\ \omega_p$, $\epsilon_2 = 3.0$, $d_2 = 0.1$, $d_3 = 0.5$ and (a) $x = 0.1$, (b) $x = 0.2$, (c) $x = 0.3$, (d) $x = 0.4$.

Fig. 1



z
ε_2
ε_1
ε_m
d_2
d_1
d_m
$2L$
L
0
d_0
$\vec{p}$

Fig. 2

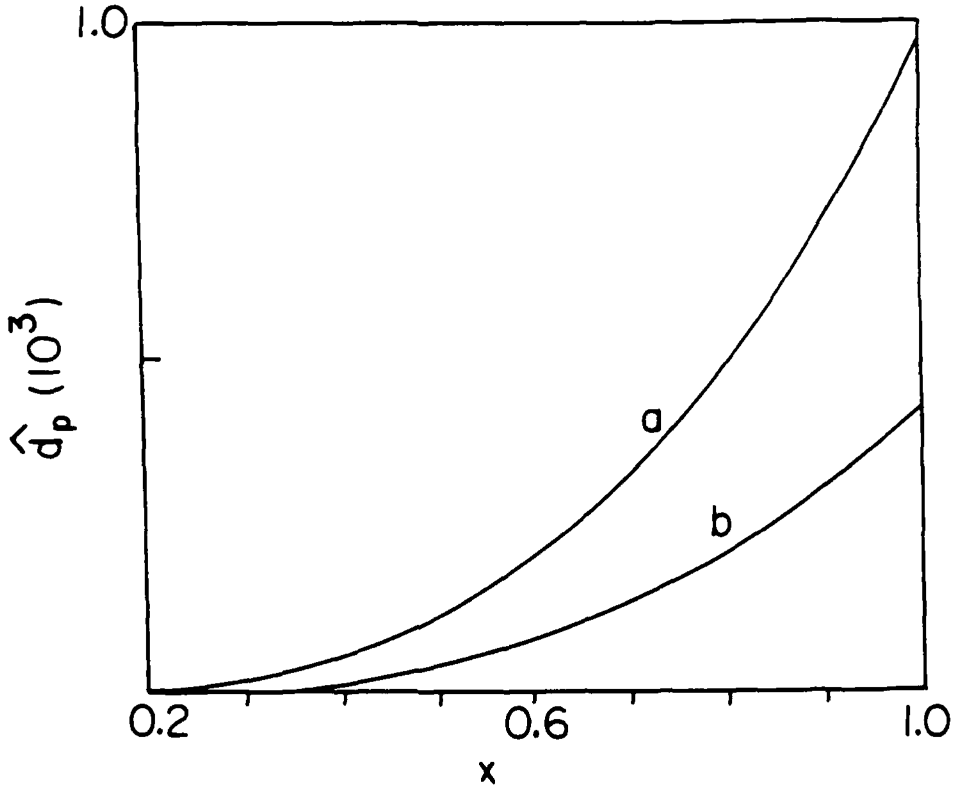
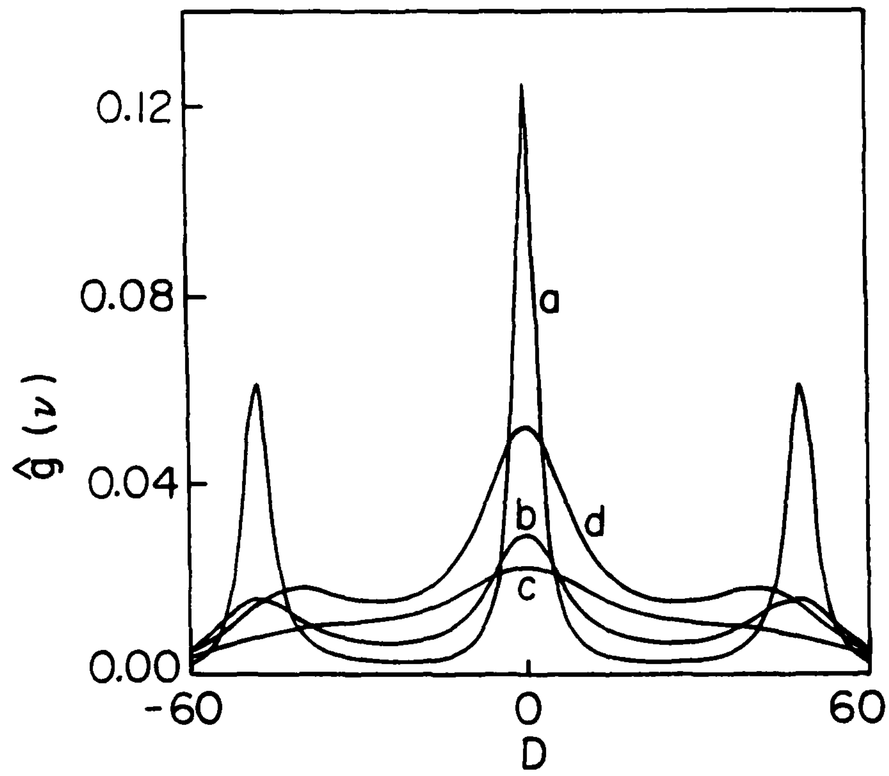


Fig. 3



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